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(54) Water repellent composition

Hydrophobiermittel

Agent imperméabilisant

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(56) References cited: US-A- 3 639 317

> PATENT ABSTRACTS OF JAPAN vol. 004, no. 101 (C-019), 19 July 1980 & JP 55 062971 A (KANSAI PAINT CO LTD), 12 May 1980.

 PATENT ABSTRACTS OF JAPAN vol. 018, no. 082 (C-1164), 10 February 1994 & JP 05 287184
 A (NIPPON PAINT CO LTD), 2 November 1993,

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Description

- [0001] This application is related to US Patent 5,421,866, issued June 6, 1995, entitled "Water Repellent Compositions" and is commonly assigned to the present proprietor.
- [0002] This invention is directed to water repellent compositions containing organosilicon compounds and polybutylene having functional groups.
 - [0003] We have unexpectedly discovered an improvement in water repellent compositions of the above mentioned patent. Therein, water repellents require as essential ingredients (i) an alkoxysilane, (ii) a silane coupling agent, (iii) polybutylene and (iv) water. The silane coupling agent is an amine or quaternary ammonium functional silane.
- [0004] Silanes (i) and (ii) are employed in a molar ratio of 0.5:1 to 3:1. The silanes are introduced as a cold blend, but are preferably first reacted with limited water, i.e., less than stoichiometric, to form a partial hydrolyzate containing an alcohol, i.e., R'OH, formed as a byproduct of the hydrolysis reaction. Thus, when alkoxysilane (i) and silane coupling agent (ii) are mixed together in the presence of water, a reaction product is formed.
- [0005] Quite to our surprise, however, is the fact that when the silane (ii) containing reactive amino or quaternary ammonium functional groups is eliminated, significant improvements are realized in water repellent effectiveness (WRE), water exclusion (WE), water repellency (WR), water absorption (WA) and beading.
 - [0006] Thus, while water repellent compositions according to our invention are similar to those of U.S. Patent 5,421,866, our water repellent compositions are "free of amine or quaternary ammonium functional silanes", which are essential therein.
- [0007] Our invention is defined in the claims and relates to a water repellent composition that is free of amine or quaternary ammonium functional silanes. This composition preferably contains 0.1-70 percent by weight of polybuty-lene; 0.1-70 percent by weight of an alkoxysilane R_nSi(OR')_{4-n}; the balance of the composition (one hundred percent by weight) being water. Preferably, the combined amount of organic polymer and alkoxysilane totals at least 3.0 percent by weight of our composition.
- [0008] The composition includes as optional components 0-70, preferably 3-70, percent by weight of an organic wax; 0-70, preferably 3-70, percent by weight of a polysiloxane; 0-20, preferably 0.1-20, percent by weight of a beading agent; 0-2, preferably 0.1-2, percent by weight of a catalyst; a solvent in an amount to provide our composition with a content of volatile organic compound (VOC) of less than 600 grams per liter; 0-10, preferably 0.1-10, percent by weight of a surfactant; and 0-1, preferably 0.1-1, percent by weight of each of a preservative; an antifoam agent; a mildewcide and a UV absorber/UV light stabilizer.
 - [0009] Our invention uses a polybutylene polymer or oligomer having a number average molecular weight (Mn) of 200-2,300, preferably less than 1,500, and most preferably less than 1,000. Such polymers and oligomers are known in the art and many are available commercially in a variety of molecular weights and end group combinations. We have found that relatively low molecular weight polybutylenes, i.e., Mn < 1,000 having terminal groups which hydrogen bond to hydroxyl groups generally found on cellulosic or masonry substrates, provide particularly superior water repellent compositions in our invention. Thus, the polybutylene polymers preferably have at least one terminal group which contains a functional group selected from epoxy, halide, alkoxyphenylene, hydroxyl, carboxyl, chlorosilyl, isocyanato, amino or amido or vinyl. A highly preferred end group is the epoxy group.
 - [0010] The alkoxysilane used herein is a single alkoxysilane or a mixture of alkoxysilanes. It has the formula R_nSi (OR')_{4-n}. R is an alkyl radical of 1-10 carbon atoms, preferably 1-6 carbons; an alkenyl radical having 2-8 carbon atoms; phenyl; chloropropyl or trifluoropropyl. n is 1 or 2; and R' is an alkyl radical having 1-6 carbon atoms. It is preferred, however, that R is a methyl or isobutyl radical and that R' is a methyl or ethyl radical.
 - [0011] Some suitable alkoxysilanes are methyltrimethoxysilane (MTMS), methyltriethoxysilane, methyltripropoxysilane, ethyltrimethoxysilane, propyltrimethoxysilane, propyltrimethoxysilane, isobutyltriethoxysilane, isobutyltriethoxysilane, dimethyldimethoxysilane, diethyldimethoxysilane, diisobutyldimethoxysilane, dibutyldimethoxysilane, dibutyldiethoxysilane and dihexyldimethoxysilane.
 - [0012] Among optional ingredients that are used according to our invention is an organic wax. The organic wax is preferably carnauba wax or a blend of petroleum and synthetic waxes. More particularly, the organic wax is a blend which includes both paraffin and polyethylene waxes. The polyethylene waxes can be high or low density polyethylene waxes or mixtures of high and low density polyethylene waxes. An exemplary organic wax and an organic wax found to be especially suitable in accordance with our invention, is JONWAX® 125, a product and trademark of SC Johnson & Sons Inc., Racine, Wisconsin. JONWAX® 125 is sold as an aqueous emulsion of polyethylene and paraffin waxes, with a solids content of thirty-five percent. Other blended paraffin and polyethylene type waxes may also be employed.
 - [0013] Another optional ingredient that is used according to our invention is a polysiloxane. One suitable polysiloxane is a silanol terminated polydimethylsiloxane of the structure HOMe₂SiO(Me₂SiO)_xSiMe₂OH where Me is methyl and x typically varies from 10 to 1,000. In general, such silanol fluids have a viscosity ranging from 15 to 20,000 centistokes (mm²/s) measured at 25°C. The terminal silanols render these polydimethylsiloxane fluids susceptible to condensation

under both mild acid and base conditions. When the end groups are exposed to moisture, a rapid crosslinking reaction takes place.

[0014] Examples of such silanol fluids are hydroxyl-terminated polydimethylsiloxanes having viscosities of 55-90 mm²/s measured at 25°C. and having a hydroxyl content of 1-2.5 weight percent; hydroxyl-terminated polydimethylsiloxanes having viscosities of 1,800-2,200 mm²/s measured at 25°C.; hydroxyl-terminated polydimethylsiloxanes having viscosities of 3,500-4,500 mm²/s measured at 25°C.; and polydimethylsiloxanes having viscosities of 11,000-14,000 mm²/s measured at 25°C., where 85-95 percent of the terminal groups are silanol and 5-15 percent of the terminal groups are trimethylsiloxy.

[0015] Another suitable polysiloxane is a silicone resin. Most preferred are solvent solutions of hydroxyl-functional organosiloxane resin copolymers essentially consisting of $CH_3SiO_{3/2}$ units, $(CH_3)_2SiO_{2/2}$ units, $C_6H_5SiO_{3/2}$ units and $(C_6H_5)_2SiO_{2/2}$ units, in a molar ratio of 25:19:37:19 and having a hydroxyl content of 0.5 percent by weight to 3.0 percent by weight.

[0016] One specific silicone resin that is useful is a 1:1 xylene: toluene solution containing 50 percent by weight solids of a hydroxyl-functional organosiloxane resin copolymer with $CH_3SiO_{3/2}$ units, $(CH_3)_2SiO_{2/2}$ units, $C_6H_5SiO_{3/2}$ units and $(C_6H_5)_2SiO_{2/2}$ units, in a molar ratio of 25:19:37:19 and having a hydroxyl content of 0.5 percent by weight.

[0017] Another suitable silicone resin that is used is a toluene solution containing 60 percent by weight solids of a hydroxyl-functional organosiloxane resin copolymer with $CH_3SiO_{3/2}$ units, $(CH_3)_2SiO_{2/2}$ units, $C_6H_5SiO_{3/2}$ units and $(C_6H_5)_2SiO_{2/2}$ units, in a molar ratio of 25:19:37:19 and having a hydroxyl content of 3.0 percent by weight.

[0018] A third specific silicone resin that is suitable is a xylene solution containing 50 percent by weight solids of a hydroxyl-functional organosiloxane resin copolymer with CH₃SiO_{3/2} units, (CH₃)₂SiO_{2/2} units, C₆H₅SiO_{3/2} units and (C₆H₅)₂SiO_{2/2} units, in a molar ratio of 25:19:37:19.

[0019] Examples of other silicone resins that are useful include organosilicon resinous copolymers including $SiO_{4/2}$ units and one or more $R_3SiO_{1/2}$ units, $R_2SiO_{2/2}$ units and $RSiO_{3/2}$ units, in a molar ratio such that the number average molecular weight of the resinous copolymer is 1,200-10,000 dalton. R is an alkyl radical with 1-6 carbon atoms; an aryl radical such as phenyl, tolyl and xylyl; an alkenyl radical such as vinyl and allyl; or a trifluoropropyl radical.

[0020] Following are three (3) specific resinous copolymers which can be used:

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I. An organic solvent soluble (i.e., preferably benzene) resin copolymer of triorganosiloxy units $R_3SiO_{1/2}$ and $SiO_{4/2}$ units in the mole ratio of 0.7 mole of triorganosiloxy units per mole of $SiO_{4/2}$ units. R has the meaning defined above. This resin has a number average molecular weight of 5,000 dalton based on gel permeation chromatography using silicate resin standards. The triorganosiloxy units are trimethylsiloxy units and dimethylvinylsiloxy units and the resin includes 1.4-2.2 weight percent silicon bonded vinyl radicals.

II. A resinous copolymeric siloxane prepared by (i) forming an acidic homogenous mixture of a silanol containing resinous copolymeric siloxane with $\rm H_3SiO_{1/2}$ units and $\rm SiO_{4/2}$ units; an organohydrogen polysiloxane with the formula $\rm R_mH_nSiO_{(4-m-n)/2}$ where m and n are positive integers with a sum less than four, preferably 1.9-2.1; and an organic solvent, and (ii) heating the mixture to remove substantially all organic solvent. R has the meaning defined above. R can also be an arylalkyl radical such as betaphenylethyl and betaphenylpropyl; or a cycloaliphatic radical such as cyclopentyl, cyclohexyl and cyclohexenyl.

III. A siloxane resin copolymer including $R_3SiO_{1/2}$ units and $SiO_{4/2}$ units in a mole ratio such that the number average molecular weight is 1,200 to 10,000 dalton. Preferably, the mole ratio is 0.7:1.0 and the number average molecular weight is 5,000. R is as previously defined. The resin contains 2.5 weight percent silicon bonded OH groups. The resin may also contain $R_2SiO_{2/2}$ units and $R_2SiO_{3/2}$ units.

[0021] Such silicone resins are described in detail in numerous patents including US Patents 2,504,388; 2,676,182; 2,706,190; 3,079,281; 4,310,678 and 4,322,518.

[0022] Another suitable polysiloxane is a solid flake nonpolar silsesquioxane resin containing 72.5 mole percent of $C_6H_5SiO_{3/2}$ units; 27.5 mole percent of $C_3H_7SiO_{3/2}$ units; and 5 percent by weight of OH groups. This type of resinous composition can be represented as $(PhSiO_{3/2})_x(PrSiO_{3/2})_yOH$ in which Ph is phenyl, Pr is propyl and the ratio of x:y is 7:3.

[0023] Another suitable polysiloxane is an aqueous silicone resin emulsion prepared by (A) hydrolyzing at least one organochlorosilane in the presence of an organic solvent to form a silicone resin hydrolyzate solution, whereby said silicone resin hydrolyzate has a residual hydrolyzable chloride content of 15-100 parts per million by weight; (B) stripping said hydrolyzate solution to reduce the organic solvent content thereof; and (C) emulsifying the solution resulting from step (B) in water with the aid of at least one anionic surfactant to form a uniform emulsion; with the proviso that the solvent content reduction according to step (B) provides an emulsion which forms a continuous film, when applied to a substrate and dried thereon.

[0024] The silicone hyrolyzate includes at least two units such as $MeSiO_{3/2}$ units, $Me_2SiO_{2/2}$ units, PhMeSiO_{2/2} units, PhSiO_{3/2} units, Ph₂SiO_{2/2} units and PrSiO_{3/2} units; where Me is methyl, Ph is phenyl and Pr is propyl, in which the

silicone has a hydroxyl content of 0.5-6.0 weight percent.

[0025] This aqueous silicone resin emulsion is more completely described in US Patent 5,300,327.

[0026] Another optional ingredient that is used in our invention is a beading agent. Representative examples are stearates such as aluminum stearate and magnesium stearate; borate salts such as sodium borate and hydrophobic silica. These materials assist in shedding films of water from a surface by forming droplets.

[0027] Another optional ingredient is a catalyst. Representative examples of catalysts are metal titanates such as dibutyltin dilaurate (DBTDL) and dibutyltin diacetate (DBTDA); acids such as acetic acid; and bases including amines such as triethanolamine (TEA), morpholine and diethylamine. Such catalysts are capable of converting alkoxysilanes to resinous products by hydrolysis and condensation.

[0028] Another optional ingredient of our invention is a solvent. Representative examples of solvents are organic solvents such as isopropanol and mineral spirits; glycol ethers such as diethylene glycol butyl ether and propylene glycol methyl ether sold under the trademark Dowanol® by The Dow Chemical Company. Midland, Michigan; and polyglycols such as ethylene glycol and propylene glycol.

[0029] Another optional ingredient used in our invention is a surfactant. Most preferred is a combination of low and high HLB nonionic surfactants. As used herein "HLB" means hydrophillic-lipophillic balance value. The low HLB nonionic surfactant has an HLB value less than 10.5, preferably less than 6.0. Representative emulsifiers in this category are: (a) BrijTM 52 which is a polyoxyethylene cetyl ether and a product of ICI Americas Inc., Wilmington, Delaware, having an HLB value of 4.9; (b) BrijTM 72 which is a polyoxyethylene stearyl ether and a product of ICI Americas Inc., Wilmington, Delaware, having an HLB value of 4.9; (c) Arlacel™ 60 which is sorbitan stearate and a product of ICI Americas Inc., Wilmington, Delaware, having an HLB value of 4.7; (d) Aldo® MS which is glycerol monostearate and a product of Lonza Inc., Fairlawn, New Jersey, having an HLB value of 3.9; (e) Aldo® PGHMS which is propylene glycol monostearate and a product of Lonza Inc., Fairlawn, New Jersey, having an HLB value of 3.0; (f) Mapeg® EGMS which is ethylene glycol monostearate and a product of PPG/Mazer, Gurnee, Illinois, having an HLB value of 2.9; (g) Hodag™ DGS which is diethylene glycol monostearate and a product of Hodag™ Chemical Corp., Skokie, Illinois, having an HLB value of 4.7; (h) Ethox™ SAM-2 which is a polyoxyethylene stearyl amine and a product of Ethox Chemicals, Inc., Greenville, South Carolina, having an HLB value of 4.9; and (i) Macol® SA-2 which is a polyoxyethylene stearyl ester and a product of PPG/Mazer, Gurnee, Illinois, having an HLB value of 4.9. Fatty alcohols such as lauryl alcohol, myristyl alcohol and cetyl alcohol, are considered nonionic surfactants with an HLB value of one and are included as a low HLB nonionic surfactant for purposes of our invention.

[0030] The high HLB nonionic surfactant has an HLB value greater than 15.0, and preferably greater than 17.0. Representative emulsifiers in this category are: (i) Brij™ 700 which is a polyoxyethylene stearyl ether and a product of ICI Americas Inc., Wilmington, Delaware, having an HLB value of 18.8; (ii) Mapeg® S-40K which is a polyoxyethylene monostearate and a product of PPG/Mazer, Gurnee, Illinois, having an HLB value of 17.2; (iii) Macol® SA-40 which is steareth-40 and a product of PPG/Mazer, Gurnee, Illinois, having an HLB value of 17.4; (iv) Triton® X-405 which is octylphenoxy polyethoxy ethanol and a product of Union Carbide Chem. & Plastics Co., Industrial Chemicals Div., Danbury, Connecticut, having an HLB value of 17.9; (v) Macol® SA-20 which is steareth-20 and a product of PPG/Mazer, Gurnee, Illinois, having an HLB value of 15.4; and (vi) Tergitol® 15-S-20 which is a C11-C15 secondary alcohol ethoxylate and a product of Union Carbide Chem. & Plastics Co., Industrial Chemicals Div., Danbury, Connecticut, having an HLB value of 16.3.

[0031] The above surfactants are merely set forth for the purpose of identifying representative emulsifiers which may be employed in accordance with our invention. Other equivalent nonionic emulsifiers may also be substituted. Thus, it would be appropriate to use, for example, (i) other alcohol ethoxylates besides Brij™ 52, Brij™ 72 and Brij™ 700; (ii) other alkylphenol ethoxylates besides Triton® X-405; (iii) other glycerol esters of fatty acids besides Aldo® MS; and (iv) other glycol esters of fatty acids besides Aldo® PGHMS and Hodag™ DGS.

5 [0032] Thus, there are included herein the nonionic surfactants such as fluorocarbon based surfactants sold under the name Fluorad™ by the 3M Company, St. Paul, Minnesota; block copolymers of ethylene oxide and propylene oxide sold under the trademark Pluronic® by BASF Corporation, Parsippany, New Jersey; fatty acid esters sold under the name Span™ by ICI Surfactants, Wilmington, Delaware; and ethoxylated fatty acid esters sold under the name Tween™ by ICI Surfactants, Wilmington, Delaware.

[0033] Certain anionic polymeric emulsifiers can be used in combination with these nonionic surfactants such as the crosslinked hydrophobically-modified polyacrylic acid polymers sold under the trademark Pemulen® by BF Goodrich, Brecksville, Ohio; and the crosslinked hydrophobically-modified polyacrylic acid copolymers sold under the trademark Carbopol® by BF Goodrich, Brecksville, Ohio. In addition, synthetic water-soluble resins such as polyvinyl alcohols (C₂H₄O)_x can be employed. If desired, an anionic surfactant may be formed in situ by including in the mixture a base such as sodium hydroxide, triethanolamine or morpholine; and a fatty acid such as stearic acid, oleic acid or a tall oil fatty acid.

[0034] Another optional ingredient of our invention is a preservative to reduce and or eliminate microbial activity in water based emulsions. Representative examples are 5-chloro-2-methyl-4-isothiazolin-3-one sold under the name

Kathon™ LX by the Rohm and Haas Co., Philadelphia, Pennsylvania; and 1-(3-chloroallyl)-3,5,7-triaza-1-azoniaada-mantane chloride sold under the trademark Dowicil® 75 by The Dow Chemical Company, Midland, Michigan.

[0035] Another optional ingredient that in our invention is an antifoam. Suitable antifoams are silicone antifoams such as silica filled polydimethylsiloxane sold by the Dow Corning Corporation, Midland, Michigan; and organic antifoams such as hydrocarbon oils sold under the trademark Advantage® by Hercules Incorporated, Wilmington, Delaware.

[0036] Another optional ingredient in our invention, where the composition is intended as an exterior coating, is a mildewcide, including materials classified as algicides, antimicrobials, bactericides, disinfectants or fungicides; that are organic or inorganic materials which reduce biological activity on a substrate. Representative examples of some suitable mildewcides include the proprietary fungicide sold under the trademark Troysan® Polyphase® P-20T by the Troy Chemical Company, East Hanover, New Jersey; diiodomethyl-p-tolylsulfone sold under the trademark Amical® by Angus Chemical Co., Buffalo Grove, Illinois; tribasic copper sulfate; and stabilized chlorine dioxide.

[0037] Another optional ingredient of our invention, where the composition is intended as an exterior coating, is a UV absorber/UV light stabilizer. Suitable UV absorber/UV light stabilizers are substituted benzotriazole and hindered amines sold under the trademark Tinuvin® by Ciba-Geigy Corporation, Hawthorne, New York.

[0038] Water repellent compositions for treating surfaces according to our invention are made by simply mixing together the various ingredients. Where a composition is desired in the form of an emulsion, it is made by (i) making one emulsion of the several ingredients; (ii) making several emulsions each containing one or more of the ingredients and subsequently combining the several emulsions; and (iii) following the procedure for (i) or (ii) and then adding some of the ingredients directly to water. These mixtures and emulsions are made using any suitable source of shear, such as a high speed stirrer, a homogenizer, sonolator, microfluidizer, TurelloTM change can mixer, RossTM mixer or EppenbachTM colloid mill. The procedure for making the mixtures and emulsions includes the direct addition of oil to water or the indirect addition of water to oil. Preferably, the particle size of the active ingredient(s) in the discontinuous or internal phase is between 0.2 micrometers (μm) to 500 micrometers (μm).

[0039] These water repellent compositions can be formulated as a concentrated emulsion having a high solids content for later dilution and direct application to a substrate; or they can be formulated as ready-to-use emulsions with low solids content for direct application to the substrate. The actual amount of water repellent composition employed will vary, depending upon the nature of the substrate being treated, but in general, it should be sufficient to provide the substrate with a coating containing 3-40 percent by weight of the solids in the water repellent composition being applied.

[0040] Substrates suitable for treatment with water repellent compositions according to our invention include cellu-

losic surfaces suitable for treatment with water repellent compositions according to our invention include cellulosic surfaces such as wood, fabric, fiber, paper and paperboard; masonry surfaces such as porous inorganic substrates including concrete, mortar, brick, stone, gypsum, stucco, terra cotta, adobe, plaster, limestone, marble, porcelain and tile; and concrete building structures.

[0041] The method of application of our water repellent composition is preferably by topical treatment or topical coating of the substrate. However, the use of these water repellent compositions can also include their incorporation directly into a substrate during its manufacture, i.e., as an additive in a paper slurry or as an ingredient in a concrete mix prior to its setting. When applied topically, for the best results, it is preferred that the substrate be treated when it is dry, but substrates may be treated when they are damp or wet.

[0042] Following are examples to illustrate our invention in more detail. In the examples, as well as in the accompanying Tables, all percentages are on a weight basis, unless indicated otherwise.

Example I - Sample Preparation

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[0043] Aqueous emulsion water repellent compositions were prepared by blending the various components at the indicated active levels shown below. In preparing these emulsions, the polybutylene and any non-water dispersible silicone component(s), were mixed until thoroughly combined, using a standard'laboratory mixer. Air driven or electric driven mixers were used, depending upon the viscosity of each blend. The surfactant(s) were incorporated into the oil phase or into the water phase, depending upon the nature of the surfactant(s). In some instances, one surfactant was incorporated into the water phase and another surfactant was incorporated into the oil phase. The oil phase and the water phase were combined with vigorous mixing, until an oil-in-water emulsion, with the desired particle size was achieved, i.e., generally 0.2 micrometers (μ m) to 500 micrometers (μ m). The remaining water was slowly incorporated into the emulsion to achieve the desired actives level, i.e., generally 3-70 percent by weight. In some embodiments, the emulsion was then blended with a wax emulsion and any other water-dispersible silicones. The resulting mixture was diluted to the desired final actives level, i.e., a concentrated emulsion having a high solids content for later dilution and direct application to a substrate or a ready-to-use emulsion with low solids content for direct application to the substrate.

[0044] While it is apparent that many different types of surfactant(s) and combinations of surfactant(s), can be employed in preparing these emulsions, nonionic surfactants Triton® X-45 and Triton® X-705 were found to be particularly

effective.

[0045] The polybutylene (PIB) used in the examples, and in the accompanying Tables, was (i) a vinyl-terminated polybutylene having a number average molecular weight of 1,340, which is a product sold under the trademark Indopol® H-300 by Amoco Chemical Company, Chicago, Illinois; (ii) a vinyl-terminated polybutylene having a number average molecular weight of 2,160, which is a product sold under the trademark Indopol® H-1500 by Amoco Chemical Company, Chicago, Illinois; and (iii) an epoxidized polybutylene having an estimated number average molecular weight of 1,000, which is a product sold under the trademark Vikopol® 64 by Elf Atochem North America, Philadelphia, Pennsylvania. [0046] The optional wax ingredient used in these examples, and referred to in the accompanying Tables, was a 35% solids emulsion of a wax consisting of 50% polyethylene and 50% paraffin. Such wax emulsions are available commercially from SC Johnson & Sons Inc., Racine, Wisconsin, as JONWAX® 125; and from Michelman Inc., Cincinnati, Ohio.

Example II - Wood

[0047] An aqueous water repellent composition for the treatment of wood, representative of our invention, was prepared according to the procedure in Example I. The composition contained on a weight basis, the following active ingredients, with the balance of the composition to 100% being water:

Γ	Amount (%)	Ingredient
Γ	2.10	PIB Indopol® H-300
	0.90	IsobutyItriethoxysilane
ı	4.00	Wax - Wax Emulsion, Michelman Inc.
1	5.00	Surfactant

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[0048] The surfactant was a mixture containing (i) a fatty acid nonionic surfactant sold under the trademark Actino® FA-3 by the Arizona Chemical Company, Panama City, Florida; (ii) triethanolamine; and (iii) Triton® X-705, a trademark of Union Carbide Chem. & Plastics Co., Industrial Chemicals Div., Danbury, Connecticut, for octylphenoxy polyethoxyethanol, a nonionic surfactant.

30 [0049] This composition did not contain an amine or quaternary ammonium functional silane.

Example III - Wood

[0050] Another aqueous water repellent composition for the treatment of wood, representative of our invention, was prepared according to the procedure in Example I. The composition contained on a weight basis, the following active ingredients, with the balance of the composition to 100% being water:

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Amount (%)	Ingredient
1.50	PIB Indopol® H-300
0.75	Isobutyltriethoxysilane
0.75	Silicone Resin - Me ₂ SiO _{2/2} and PhMeSiO _{2/2} units
4.00	Wax - Wax Emulsion, Michelman Inc.
5.00	Surfactant
0.75 0.75 4.00	IsobutyItriethoxysilane Silicone Resin - Me ₂ SiO _{2/2} and PhMeSiO _{2/2} units Wax - Wax Emulsion, Michelman Inc.

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[0051] The surfactant mixture used in Example II was used in this example. This composition also did not contain an amine or quaternary ammonium functional silane.

Example IV - Comparison - Wood

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[0052] An aqueous water repellent composition for the treatment of wood, NOT representative of our invention, was prepared according to the procedure in Example I. The composition contained on a weight basis, the following active ingredients, with the balance of the composition to 100% being water:

Amount (%)	Ingredient
2.51	PIB Indopol® H-300

(continued)

	Amount (%)	Ingredient
	1.24	Isobutyltriethoxysilane
5	1.25	A mixture of water, methyltrimethoxysilane and N-(2-aminoethyl)-3-aminopropyltrimethoxysilane
	5.00	Surfactant - Tergitol® TMN-6

[0053] This composition DID contain an amine or quaternary ammonium functional silane, i.e., N-(2-aminoethyl)-3-aminopropyl trimethoxysilane. It is representative of water repellent compositions taught by US Patent 5,421,866, over which our invention is an improvement.

Example V - Comparison - Wood

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[0054] Another aqueous water repellent composition for the treatment of wood, NOT representative of our invention, was prepared according to the procedure in Example I. The composition contained on a weight basis, the following active ingredients, with the balance of the composition to 100% being water:

Amount (%)	Ingredient	
2.50	PIB Indopol® H-300	
2.50	A mixture of water, methyltrimethoxysilane and N-(2-aminoethyl)-3-aminopropyltrimethoxysilane	١
5.00	Surfactant - Tergitol® TMN-6	

[0055] This composition DID contain an amine or quaternary ammonium functional silane, i.e., N-(2-aminoethyl)-3-aminopropyl trimethoxysilane. It is also representative of water repellent compositions taught by US Patent 5,421,866, over which our invention is an improvement.

[0056] The water repellent compositions prepared according to Examples II-V were used to treat wood and evaluated for water repellency, according to three industry standard primary test methods. The three methods used were (i) The Swellometer Test Method according to Federal Specification TT-W-572B and ASTM D4446; (ii) The Gravimetric Test Method according to ASTM D5401; and (iii) The Beading Test Method.

[0057] The Swellometer Test Method- Fed. Spec. TT-W-572B or ASTM D4446

[0058] This test evaluates protection provided to wood by various treatments, by measuring both water pick-up and dimensional stability of treated versus untreated wood samples. The wood samples were $6.4 \times 38.1 \times 254$ cm (0.25 x 1.5 x 10 inches) cross-sectional wafers, cut from straight-grained, knot-free, flat-sawn, clear, average density, kilndried, Southern Pine sapwood. All wood wafers were conditioned at $50 \pm 5\%$ relative humidity and $70 \pm 5\%$. (21 $\pm 3\%$ C.) until a constant weight (± 0.5 g in 24 hours) and moisture content (12.5%) was reached. Moisture content was determined by ASTM D4442. The wood was dried in a 217 $\pm 5\%$ F. (103 $\pm 2\%$ C.) oven and moisture content (MC) was calculated by the formula:

$$\% MC = \frac{\text{(original mass - oven-dry mass) x 100}}{\text{oven-dry mass}}$$

[0059] Wafers were removed from the conditioning room and immediately treated by soaking 3 minutes in water-based formulations or 30 seconds in solvent-based formulations. Treated wafers were then placed on a raised screen or rack at a 45° angle and allowed to air dry at ambient conditions overnight. An untreated wafer taken from consecutive pieces of the same board was used as a control for each treated wafer. Sets of treated and untreated pairs were then placed in a chamber and conditioned at $65 \pm 5\%$ relative humidity and 70 ± 5 °F. (21 \pm 3°C.) until constant weight was reached (7 days).

[0060] Wafers were placed in a swellometer test apparatus, which was then submerged in deionized water maintained at 75 \pm 5°F. (24 \pm 3°C.) for 30 minutes. The dimensional change of the wafer was measured by the swellometer gauge during the soak and was used to calculate % Water Repellency (WR). After removal from the gauge, the wood was then weighed and the % Water Exclusion (WE) was determined from the degree of weight gain.

[0061] The percent water repellency (% WR) was calculated by comparing the swell of the treated wafer to that of the untreated wafer with the formula:

[0062] The percent water exclusion (% WE) was calculated by comparing the weight gain of the treated wafer to the untreated wafer with the formula:

The Gravimetric Test Method according to ASTM D5401

[0063] The gravimetric test was used to evaluate the effectiveness of clear water repellent coatings on wood. The substrates were $5.1 \times 10.2 \times 15.2$ cm. (2 x 4 x 6 inches) straight-grained, knot-free, flat-sawn, kiln-dried, Ponderosa Pine sapwood boards, conditioned at $50 \pm 5\%$ relative humidity and $70 \pm 5\%$. (21 $\pm 3\%$.) until a constant weight (± 1 g in 24 hours) and moisture content (12.5%) was reached. Moisture content was determined by ASTM D4442 in the same manner as above.

[0064] After these conditions were met, the boards were removed from the conditioning room and immediately treated by soaking 3 minutes in water-based formulations or 30 seconds in solvent-based formulations. The treated boards were placed on a raised rack and left to cure in the laboratory at ambient conditions overnight. Three control boards for each day of testing were left untreated and kept in the 50 % humidity room. Boards were then equilibrated in the 50 % humidity room until weight change in two successive 24 hour measurements was less than 0.5 g (6-7 days).

[0065] The treated and untreated boards were each weighed and then allowed to float in distilled/deionized water maintained at 75 ± 5 °F. (24 \pm 3°C.) for 30 minutes, 15 minutes on each side, turning them over in between. The boards were removed, the excess water was wiped off and each was re-weighed.

[0066] The percent Water Repellent Effectiveness (% WRE) was then calculated by comparing the average weight gain of the treated boards, to the average weight gain of the untreated controls, with the formula:

The Beading Test Method

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[0067] The beading test was used to evaluate the beading ability of various samples on wood. The beading for each composition was tested using the gravimetric boards described above prior to their testing. Approximately 6-8 water droplets were dripped onto the board using an eye-dropper and then the boards were sprayed with water. Beading was evaluated on a scale from 0-5, with 5 being exceptional beading with tight spherical beads, 3 being moderate beading with flatter irregular shaped beads and 0 being no beading.

[0068] Table I shows the results on wood for the water repellent compositions of Examples II-V in evaluations according to (i) The Swellometer Test Method, (ii) The Gravimetric Test Method, and (iii) The Beading Test Method.

TABLE I

Example	Gravimetric % WRE	Swellometer % WE	Swellometer % WR	Beading 0-5
II	86	77	67	4+
III	88	78	70	4+
IV	61	6	1	3+
V	72	14	3	4

[0069] Water repellent compositions according to our invention in Examples II and III, were much more effective in all categories evaluated, as is seen in Table I, in comparison to water repellent compositions NOT according to our invention, shown in Examples IV and V. In particular, note that the values for % Water Exclusion (WE) and % Water Repellency (WR) varied as much as 10-20 fold or more.

Example VI - Sandstone

[0070] An aqueous water repellent composition for the treatment of sandstone, representative of our invention, was prepared according to the procedure in Example I. The composition contained on a weight basis, the following active ingredients, with the balance of the composition to 100% being water:

Amount (%)	Ingredient
1.00	PIB Indopol® H-300
1.00	PIB Indopol® H-1500
1.00	IsobutyItriethoxysilane
4.00	Wax - Wax Emulsion, Michelman Inc.
5.00	Surfactant

[0071] The surfactant was the same mixture used in Examples II and III, containing (i) Actinol® FA-3; (ii) trieth-anolamine; and (iii) Triton® X-705.

[0072] This composition did not contain an amine or quaternary ammonium functional silane.

Example VII - Comparison - Sandstone

[0073] An aqueous water repellent composition for the treatment of sandstone, NOT representative of our invention, was prepared according to the procedure in Example I. This composition contained on a weight basis, the same active ingredients used in Example V, i.e., PIB Indopol® H-300; methyltrimethoxysilane; N-(2-aminoethyl)-3-aminopropyltrimethoxysilane; Tergitol® TMN-6 and water.

[0074] This composition DID contain an amine or quaternary ammonium functional silane, i.e., N-(2-aminoethyl)-3-aminopropyl trimethoxysilane. It is representative of water repellent compositions taught by US Patent 5,421,866, over which our invention is an improvement.

Example VIII - Mortar

[0075] An aqueous water repellent composition for the treatment of mortar, representative of our invention, was prepared according to the procedure in Example I. The composition contained on a weight basis, the same active ingredients used in Example VI, i.e., PIB Indopol® H-300; PIB Indopol® H-1500; isobutyltriethoxysilane; Michelman Inc., Wax Emulsion; and the surfactant was the same mixture used in Examples II, III and VI, containing (i) Actinol® FA-3; (ii) triethanolamine; and (iii) Triton® X-705.

[0076] This composition did not contain an amine or quaternary ammonium functional silane.

Example IX - Mortar

[0077] Another aqueous water repellent composition for the treatment of mortar, representative of our invention, was prepared according to the procedure in Example I. The composition contained on a weight basis, the following active ingredients, with the balance of the composition to 100% being water:

Amount (%)	Ingredient
28.00	PIB Indopol® H-300
10.00	IsobutyItriethoxysilane
40.00	Silicone Resin - Me ₂ SiO _{2/2} and PhMeSiO _{2/2} units
5.00	Surfactant

[0078] The surfactant was the same mixture used in Examples II, III, VI and VIII, containing (i) Actinol® FA-3; (ii) triethanolamine; and (iii) Triton® X-705. This composition also did not contain an amine or quaternary ammonium functional silane.

Example X - Comparison - Mortar

[0079] An aqueous water repellent composition for the treatment of mortar, NOT representative of our invention, was prepared according to the procedure in Example I. This composition contained on a weight basis, the same active ingredients used in Examples V and VII, i.e., PIB Indopo® H-300; methyltrimethoxysilane; N-(2-aminoethyl)-3-amino-propyl trimethoxysilane; Tergitol® TMN-6 and water.

[0080] This composition DID contain an amine or quaternary ammonium functional silane, i.e., N-(2-aminoethyl)-3-aminopropyl trimethoxysilane. It is representative of water repellent compositions taught by US Patent 5,421,866,

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over which our invention is an improvement.

[0081] The water repellent compositions prepared according to Examples VI and VII were used to treat sandstone and the water repellent compositions prepared according to Examples VIII-X were used to treat mortar and evaluated for water absorption (WA), water exclusion (WE) and beading, according to industry standard primary test methods. The methods used were (i) The Federal Specification SS-W-110C, a test procedure for masonry, which covers sandstone and mortar; and (ii) The Beading Test Method referred to above.

Test Procedure - Masonry - Federal Specification SS-W-110C

[0082] This specification covers clear penetrating solutions which provide water repellency to exterior masonry materials such as sandstone and mortar. The water absorption (WA) characteristics of a substrate are evaluated by soaking each block in 6.4 mm (1/4 inch) of water for three days. The results are reported as a percent water absorption (% WA) based on dry weight of the cube and as percent water exclusion (% WE) based on difference in water uptake between treated and untreated cubes.

[0083] Duplicate substrates were treated for each emulsion prepared in Examples VI-X. Sandstone cubes were cut from Briar Hill sandstone to the dimensions of 2.5 x 2.5 x 10.2 cm (1 x 1 x 4 inches). Mortar cubes were made with Type I cement cured 28 days and cut into 5.1 cm. (2 inch) cubes. The face of each substrate was brushed with a wire brush and the excess dust was blown off with air. The cubes were dried in an oven at 80°C. ± 5°C. until they reached a constant weight, i.e., varying less than 0.2 grams over a four hour period. After the cubes had reached a constant weight, they were allowed to cool to room temperature. The weight of each sandstone and mortar cube was determined within 0.1 g and recorded.

[0084] Untreated control cubes were placed in a 6.4 mm (1/4 inch) of water for 24 hours. Then the cubes were removed from the water and excess water removed with a damp cloth. The wet weights were determined and the amount of water absorbed was calculated as a percent of dry weight. The cubes were returned to the oven to dry until they reached a constant weight.

[0085] Once the untreated cubes had reached a constant weight, they were allowed to cool to room temperature, weighed and immediately treated with the test composition for 30 seconds. The amount of coverage was determined by the difference of dry weight to wet weight, i.e., 151/grams of treatment = $6.0132 \text{ m}^2/\text{m}^3$ (151/grams of treatment x $4.07 = X \text{ ft}^2/\text{gal}$). The treated cubes were placed on a rack in the laboratory and rotated every minute for the first hour. The cubes were allowed to cure at room temperature for 48 hours.

[0086] After 48 hours, the weight of each cube was determined and the cubes were immersed in a 6.4 mm (1/4 inch) of water for 72 hours. The appearance of each cube was noted, as well as the substrate ability to bead water, prior to the cubes placement in water. The cubes were removed after 72 hours and the excess water was removed with a cloth. The cubes were weighed and the amount of water absorbed was calculated based on the dry weight of the cube.

35 Calculations were made using the formulas:

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where C1 is the uncoated substrate before immersion; C2 is the uncoated substrate after immersion; S1 is the treated substrate before immersion; and S2 is the treated substrate after immersion.

[0087] Table II shows the results on sandstone for the water repellent compositions of Examples VI and VII, in evaluations according to (i) Federal Specification SS-W-110C Test Procedure for Masonry, and (ii) The Beading Test Method.

TABLE II

Example	SS-W-110C % WA	SS-W-110C % WE	Beading 0-5
VI	0.6	92	4
VII	8.8	-20	0

[0088] Water repellent compositions according to our invention in Example VI, were again much more effective in all categories evaluated, as seen in Table II, in comparison to water repellent compositions NOT according to our invention, shown in Example VII. In particular, note that the values for % Water Absorption (WA) and % Water Exclusion

(WE) varied significantly and that beading was much improved.

[0089] Table III shows the results on mortar for the water repellent compositions of Examples VIII-X, in evaluations according to (i) Federal Specification SS-W-110C Test Procedure for Masonry, and (ii) The Beading Test Method.

TABLE III

Example	SS-W-110C % WA	SS-W-110C % WE	Beading 0-5
VIII	1.6	80	1+
IX	0.4	95	1
X	7.9	11	2

[0090] Water repellent compositions of our invention in Examples VIII and IX, were again much more effective, as seen in Table III, in comparison to water repellent compositions NOT according to our invention, shown in Example X. In particular, note that the values for % Water Exclusion (WE) varied significantly.

Claims

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- 20 1. A composition comprising a mixture formed by combining water; a polybutylene polymer or oligomer having a number average molecular weight of 200-2,300 and at least one terminal group selected from vinyl, epoxy, halide, alkoxyphenylene, hydroxyl, carboxyl, chlorosilyl, isocyanato, amino and amido; and an alkoxysilane of the formula R_nSi(OR')_{4-n} where R is an alkyl radical of 1-10 carbon atoms, an alkenyl radical having 2-8 carbon atoms, phenyl, chloropropyl or trifluoropropyl, n is 1 or 2 and R' is an alkyl radical having 1-6 carbon atoms; the composition being free of amine or quaternary ammonium functional silanes.
 - 2. A composition according to claim 1 in which the alkoxysilane is selected from the group consisting of methyltrimethoxysilane, methyltripropoxysilane, ethyltrimethoxysilane, ethyltributoxysilane, propyltrimethoxysilane, propyltriethoxysilane, isobutyltrimethoxysilane, butyltriethoxysilane, hexyltrimethoxysilane, dimethyldimethoxysilane, diethyldimethoxysilane, diisobutyldimethoxysilane, ysilane, phenyltrimethoxysilane, dibutyldiethoxysilane and dihexyldimethoxysilane.
 - 3. A composition according to claim 1, in which the polybutylene is vinyl-terminated and has a number average molecular weight of 1340 or 2160.
 - 4. A composition according to claim 1, in which the polybutylene has an epoxy end group.
 - 5. A composition according to claim 1 further comprising at least one additional component selected from the group consisting of an organic wax, a polysiloxane, a beading agent, a catalyst, a solvent in an amount to provide the composition with a content of volatile organic compounds less than 600 grams per liter, a surfactant, a preservative, an antifoam agent, a mildewcide and a UV absorber/UV light stabilizer.
 - 6. A composition according to claim 5 in which the additional component is a polysiloxane which is a silicone resin or silanol terminated polydimethylsiloxane.
 - 7. A composition according to claim 5 in which the additional component is at least two surfactants, one surfactant having an hydrophillic-lipophillic balance value of less than 10.5 and the other surfactant having an hydrophillic-lipophillic balance value of more than 15.
- 8. A composition according to claim 7 in which the composition is emulsified.
 - A method of rendering a cellulosic or masonry surface water repellent comprising applying to said surface the composition of claim 1.

Patentansprüche

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- 1. Zusammensetzung enthaltend eine Mischung, die gebildet wird durch Vereinigen von Wasser, einem Polybuty-lenpolymer oder -oligomer mit einem zahlenmittleren Molekulargewicht von 200 bis 2.300 und mindestens einer endständigen Gruppe, ausgewählt aus Vinyl, Epoxy, Halogenid, Alkoxyphenylen, Hydroxyl, Carboxyl, Chlorsilyl, Isocyanat, Amino und Amido, und eines Alkoxysilans der Formel R_nSi(OR')_{4-n}, worin R ein Alkylrest mit 1 bis 10 Kohlenstoffatomen, ein Alkenylrest mit 2 bis 8 Kohlenstoffatomen, Phenyl, Chlorpropyl oder Trifluorpropyl ist, n gleich 1 oder 2 ist und R' ein Alkylrest mit 1 bis 6 Kohlenstoffatomen ist, wobei die Zusammensetzung frei von Silanen mit Aminfunktionalität oder quartärer Ammoniumfunktionalität ist.
- 2. Zusammensetzung nach Anspruch 1, in der das Alkoxysilan ausgewählt ist aus der Gruppe bestehend aus Methyltrimethoxysilan, Methyltriethoxysilan, Methyltripropoxysilan, Ethyltrimethoxysilan, Ethyltributoxysilan, Propyltrimethoxysilan, Propyltrimethoxysilan, Isobutyltrimethoxysilan, Butyltriethoxysilan, Hexyltrimethoxysilan, Dimethyldimethoxysilan, Diethyldimethoxysilan, Disobutyldimethoxysilan, Diethyldimethoxysilan, Dibutyldiethoxysilan und Dihexyldimethoxysilan.
- 3. Zusammensetzung nach Anspruch 1, in der das Polybutylen eine endständige Vinylgruppe aufweist und ein zahlenmittleres Molekulargewicht von 1.340 oder 2.160 hat.
- Zusammensetzung nach Anspruch 1, in der das Polybutylen eine endständige Epoxygruppe aufweist.
 - 5. Zusammensetzung nach Anspruch 1, die weiterhin mindestens eine zusätzliche Komponente, ausgewählt aus der Gruppe bestehend aus einem organischen Wachs, einem Polysiloxan, einem Abperlmittel, einem Katalysator, einem Lösungsmittel in einer Menge, um der Zusammensetzung einen Gehalt an flüchtigen organischen Verbindungen von weniger als 600 g/l zu verleihen, einem oberflächenaktiven Stoff, einem Konservierungsstoff, einem Antischaummittel, einem Antischimmelmittel und einem UV-Absorber/UV-Lichtstabilisator, enthält.
 - 6. Zusammensetzung nach Anspruch 5, in der die zusätzliche Komponente ein Polysiloxan ist, das ein Siliconharz oder ein Polydimethylsiloxan mit Silanolendgruppen ist.
 - 7. Zusammensetzung nach Anspruch 5, in der die zusätzliche Komponente mindestens zwei oberflächenaktive Substanzen sind, wobei eine oberflächenaktive Substanz einen hydrophil/lipophil-Verhältniswert von weniger als 10,5 und die andere oberflächenaktive Substanz einen hydrophil/lipophil-Verhältniswert von mehr als 15 aufweist.
- 35 8. Zusammensetzung nach Anspruch 7, in der die Zusammensetzung emulgiert ist.
 - Verfahren, um eine zellstoffhaltige Oberfläche oder Oberfläche eines Baustoffs wasserabstoßend zu machen, umfassend Auftragen der Zusammensetzung nach Anspruch 1 auf diese Oberfläche.

Revendications

- 1. Composition comprenant un mélange formé en combinant de l'eau ; un polymère ou un oligomère de polybutylène présentant une masse moléculaire moyenne en nombre de 200 2300 et au moins un groupe terminal choisi parmi les groupes vinyle, époxy, halogénure, alcoxyphénylène, hydroxyle, carboxyle, chlorosilyle, isocyanato, amino et amido ; et un alcoxysilane de formule R_nSi (OR')_{4-n}, dans laquelle R est un radical alkyle de 1 à 10 atomes de carbone, un radical alcényle de 2 à 8 atomes de carbone, phényle, chloropropyle ou trifluoropropyle, n est 1 ou 2 et R' est un radical alkyle de 1 à 6 atomes de carbone ; la compositon étant exempte de silanes à fonctionnalité amine ou ammonium quaternaire.
- 2. Composition selon la revendication 1 dans laquelle l'alcoxysilane est choisi dans le groupe constitué de méthyltriméthoxysilane, méthytriéthoxysilane, méthyltripropoxysilane, éthyltriméthoxysilane, éthyltributoxysilane, propyltriméthoxysilane, propyltriméthoxysilane, propyltriéthoxysilane, isobutylméthoxysilane, isobutyltriéthoxysilane, butyltriéthoxysilane, hexytriméthoxysilane, diméthyldiméthoxysilane, diméthyldiéthoxysilane, diéthyldiméthoxysilane, disobutyldiméthoxysilane, phényltriméthoxysilane, dibutyldiéthoxysilane et dihexyldiméthoxysilane.
- Composition selon la revendication 1, dans laquelle le polybutylène est à terminaison vinyle et a une masse moléculaire moyenne en nombre de 1340 ou 2160.

- 4. Composition selon la revendication 1, dans laquelle le polybutylène possède un groupe d'extrémité époxy.
- 5. Composition selon la revendication 1, comprenant en outre au moins un composant supplémentaire choisi parmi le groupe constitué d'une cire organique, un polysiloxane, un agent de formation de billes, un catalyseur, un solvant en une quantité conférant à la composition une teneur en composés organiques volatils inférieure à 600 grammes par litre, un tensioactif, un agent conservateur, un agent anti-mousse, un agent antimoisissure et un absorbeur d'UV/photo-stabilisant vis-à-vis des UV.
- 6. Composition selon la revendication 5, dans laquelle le composant supplémentaire est un polysiloxane qui est une résine de silicone ou un polydiméthylsiloxane à terminaison silanol.
 - 7. Composition selon la revendication 5, dans laquelle le composant supplémentaire est constitué d'au moins deux tensioactifs, un tensioactif ayant un indice d'équilibre hydrophile-lipophile inférieur à 10,5 et l'autre tensioactif ayant un indice d'équilibre hydrophile-lipophile de plus de 15.
 - 8. Composition selon la revendication 7, dans laquelle la composition est émulsionnée.

9. Procédé pour rendre une surface cellulosique ou de maçonnerie étanche à l'eau, comprenant l'application sur ladite surface de la composition de la revendication 1.